DOE RADIONUCLIDE MONITORING SYSTEMS FOR CTBT VERIFICATION

R. W. Perkins
H. S. Miley, and W. K. Hensley

Pacific Northwest Laboratory
Richland, Washington

Contract No. DE-AC06-76RLO 1830
Sponsored by the United States Department of Energy
Office of National Security and Nonproliferation

August 1995

ABSTRACT

Radionuclide monitoring systems which could be used in monitoring a Comprehensive Test Ban Treaty (CTBT) are being developed and tested at the Department of Energy (DOE) Pacific Northwest Laboratory (PNL). One system is designed to permit automatic near real-time analysis of particulate airborne fission products with a sensitivity of about 2 $\mu Bq/m^3$ of air for the more important signature radionuclides. The second system is designed to measure the noble gas fission products Xe-133 (5.243 day) and Xe-135 (9.10 hour) with sensitivities of about 20 $\mu Bq/m^3$ of air. These systems are designed to operate automatically without maintenance for periods of months to one year or more. A ruggedized model of the particulate radionuclide monitoring system is currently undergoing field test operations at the McClellan Air Force Base. Field tests of the xenon radionuclide monitoring system are planned for early 1996. Both systems provide high sensitivity based on high flow rates and the radioxenon system achieves much high sensitivity than other reported systems based on beta coincidence gamma-ray spectrometric analysis.

Key Words: Radionuclides, Real-Time Monitoring, CTBT Verification

OBJECTIVE

The major objective of the R&D efforts are to develop and demonstrate near real-time monitoring technologies for airborne signature radionuclides which are indicative of nuclear weapons testing in any of the earth's environments. Since the beginning of the nuclear era, analytical technologies have been developed for measuring radionuclides in the atmosphere and in essentially all of the earth's environments. Many of these analytical procedures could be applied directly in monitoring for compliance to a Comprehensive Test Ban Treaty (CTBT); however, they have not been optimized for the spectrum of radionuclide of concern and their labor-intensive nature could impose prohibitive costs and unacceptable delays in data receipt for an International Monitoring System. The DOE focus in developing monitoring technology has been to design systems which could automatically and continuously monitor for those airborne signature radionuclides which provided the best indication of nuclear detonations and which also provide the highest practical sensitivity. The overall objectives have therefore included (1) the identification of the most easily observable and unique radionuclides which would be released from nuclear tests in all of the earth's environments, (2) the selection of practical measurement technologies which were adaptable to near real-time analysis and automation, and (3) the design, construction and testing of practical ruggedized monitoring systems which could operate maintenance-free for periods of months to a year or more, and transmit airborne radionuclide concentrations to National and International Data Centers, as required, at any desired frequency.

RESEARCH ACCOMPLISHED

The goals of this R&D program have been (1) to determine which radionuclides may enter the atmosphere from various nuclear test scenarios, (2) to determine their uniqueness based on other possible sources, (3) to evaluate various measurement alternatives which may be adaptable for automatic near real-time analysis, and (4) to design, construct and test monitoring systems.

A review of signatures from past nuclear detonation monitoring indicates that for ground surface and atmospheric detonation, the full spectrum of fission products enter the atmosphere; however, where underground or underwater detonation are conducted the noble gas radionuclides, and to a much lesser degree, the radioiodines are most likely to be vented.

Thus, in monitoring for compliance to a CTBT, the instrumentation should be capable of measuring the more abundant short-lived fission products from an atmospheric detonation and the more abundant noble gases from a subsurface detonation. The abundance of these radionuclides are summarized in Table 1 for periods of one day, three days, 10 days and 30 days post detonations. The ambient atmospheric concentrations of the shorter lived particulate radionuclides in Table 1 are much lower than the best current detection capabilities of about 1 $\mu Bq/m^3$ and therefore do not present a background interference to their monitoring. However, the xenon radionuclides, Xe-133 (5.243d) and Xe-135 (9.10 hour) are released to some degree from all operating nuclear reactors and therefore may limit their sensitivities for detecting nuclear detonations. The atmospheric concentration of Xe-133 in the northern hemisphere at mid-latitudes and at a few hundred kilometers from operating nuclear reactors appears to be

about 300 $\mu Bq/m^3$, whereas backgrounds at higher and lower latitudes in the northern hemisphere are one or two orders of magnitude less and those in the southern hemisphere are at least two orders of magnitude less. The measurement sensitivity of the DOE monitoring system with its sensitivity of about 20 $\mu Bq/m^3$ should therefore be capable of continuously monitoring the Xe-133 at mid-latitudes in the northern hemisphere and detect significant changes in these concentrations. It should also be capable of detecting concentrations in excess of 20 μBq in the southern hemisphere which would presumably be due to a nuclear detonation or a major nuclear reactor release. The ambient concentrations of the shorter level Xe-135 (9.10 ks) appears to be at least 50-fold lower and its observation at high levels would be suggestive of debris from a nuclear detonation.

The quantities of fission product radionuclides which are produced in even a small (1 Kt) nuclear detonation are considerable, and if such a test were conducted in the atmosphere it should be readily detectable within a few days based on proposed 100 station particulate and noble gas radionuclide monitoring arrays.

Table 2 shows the quantities of two of the major particulate radionuclides, Mo-99 (2.75d) and Ba-140 (12.75d), and the two major noble gas radionuclides Xe-133 (5.243d) and Xe-135 (9.10 hour) which would result from the detonation of a 1 Kt plutonium fission device at one day post-detonation. It also shows what these average concentrations would be if they were diluted by the entire earth's atmosphere and compares their concentrations with the sensitivities which are achievable with the DOE monitoring systems which are being developed and tested. It is clear that atmospheric tests, where 100% of the debris enters the atmosphere, should not be difficult to detect.

Whether denotations occur underground or underwater, only a very small fraction of the total fission products may enter the atmosphere and be transported by turbulent mixing, convective processes and the prevailing winds. In such cases, it is essential to have monitoring systems which provide the highest practical sensitivity. Also, any vented material would be highly enriched in the noble gas radionuclides, and these would be present in ratios proportioned to their independent fission fields which are very different from the "chain yields" which would be generated from precursor radionuclide decay in an atmospheric detonation. The quantities of xenon radionuclides which would be present during the first few minutes following a 1 Kt nuclear plutonium detonation and their ratios on release and during atmospheric transport are shown in Table 3. For an enriched uranium weapon the ratios would be about 1/5 of these values. The ability to measure both the Xe-133 and Xe-135 is thus important since it could serve as a basis for confirming the detonation time of the event where the fissile material was known (plutonium or enriched uranium) or to determine the type of device where the detonation time was known for subsurface tests.

Based on the above factors and other considerations, near real-time and fully automatic monitoring systems are being developed for the continuous measurement of particulate and xenon radionuclides which are indicative of nuclear detonation.

Particulate Radionuclide Analyzer

A particulate radionuclide analyzer which is capable of continuously separating particulate radionuclides from the atmosphere over any specified time period followed by their automatic measurement and transmission of gamma-ray spectral data and specified radionuclide concentrations has been developed and various prototype models have been tested over the last eight months. The initial two prototypes were operated in a connex container near our laboratory at Richland, Washington while the third prototype is currently being tested at the McClellan Air Force Base in Sacramento, California.

These automated particulate radionuclide analyzer has been developed and tested over the past two years. The analyzer draws large volumes of air (15 to 25 m^3/min) through a large area (0.25 m^2), low pressure-drop filter that is subsequently mechanically folded, sealed in a plastic-sieve bag, then positioned at the face of a large-volume, high-resolution germanium detector for gamma-ray spectrometric analysis. The analyzer is equipped to analyze the resulting spectra, transmit the data to a central data repository, accept dial-in commands to perform calibrations, change the sampling schedule, perform various diagnostics, and access stored information and associated information including meteorological data, historical detector performance data, historical system parameters, and all past and present gamma-ray spectral information. During these dial-up sessions, this system can also perform remote software upgrades and do limited mechanical maintenance and repairs. This interrogation/command process can be conducted by network or phone link (hardwired or satcom) using point-to-point protocol.

The product of our design and development efforts is a particulate radionuclide analyzer which consumes about 2500W of power, can process approximately 540 samples between reloading, is about 90 cm x 210 cm, and should cost roughly $100K. Measurement sensitivities for the principal fission products of interest to the CTBT community are about 2 μBq/SCM. Coupled with the expected radionuclide inventory of a 1 Kt detonation and the proposed 100 station world-wide sampling network, any surface or atmospheric nuclear testing should be readily observable. Photographs of the particulate radionuclide analyzer are shown in Figure 1 and include a summary of its operating parameters together with the current joint field test setups at the McClellan Air Force Base.

Xenon Radionuclide Analyzer

The automated xenon radionuclide analyzer is designed to continuously separate the Xe-133 (5.243d) and Xe-135 (9.10 ks) from the atmosphere during eight hours (or other selected) time intervals, measure these radionuclides by low background beta coincidence gamma-ray spectrometry and automatically transmit the gamma-ray spectra and xenon radionuclide concentrations to the National and International Data Centers. The combination of large volume air sampling (40 m^3 /8 hour), the extremely efficient beta coincidence gamma-ray counting (70 to 80%), the very low counter background and the long counting periods (up to 32 hour) permits reliable measurement of both Xe-133 and Xe-135 at concentrations as low as 20 μBq/m^3 of air.

The xenon concentration and purification process involves passing filtered air through a mixed bed of molecular sieve and aluminum oxide for removal of water, carbon dioxide and parts of

the radon. The air stream then passes through a refrigerator where it is cooled to a -100°C and then through an activated charcoal sorption bed maintained at the same temperature for eight hours (or other selected) time periods. At the end of the sampling period, the xenon is thermally desorbed, recaptured and purified on a much smaller sorption bed from which it is transferred to the beta counting cell for beta-coincidence gamma-ray spectrometric analysis. This counting system shown in Figure 2 allows four samples to be measured simultaneously and thus permits continuous eight-hour collections and up to 32-hour counting periods.

RECOMMENDATIONS AND FUTURE PLANS

Both the particulate and xenon radionuclide analyzers are under development, and this will continue together with test programs during the next year and beyond (Figure 3). The particulate radionuclide system is further along in its development and the current ruggedized field version could probably be employed in an actual monitoring operation. Following its testing at the McClellan Air Force Base, and any modifications dictated by these tests, the system will be operated near nuclear reactors such that actual fission product signatures may be observed. Also, optimization studies will be conducted to determine the most practical time intervals for sample collections, sample decay (if any), and counting periods.

Similar tests are planned for the xenon radionuclide analyzer. However, these will be approximately one year later than those planned for the particulate radionuclide analyzer.

We are currently discussing these instrument designs with commercial firms with the objective of having then commercially available for possible use in monitoring a CTBT or for other monitoring applications around nuclear facilities and elsewhere. It is currently planned that the particulate and xenon radionuclide monitoring systems will be commercially available in January and December of 1997, respectively.

Table 1

PRINCIPAL REMAINING FISSION PRODUCTS FROM A 1 kT BURST*
AFTER 1 to 30-DAY DECAY PERIODS (Gamma-ray Emitters)

1-DAY DECAY			3-DAY DECAY		
Radionuclide	**Half-Life**	**%**	**Radionuclide**	**Half-Life**	**%**
			Mo-99	2.75 d	9.6
Xe-135	9.089 hr	11.8	Rh-105	1.473 d	8.3
I-133	20.8 hr	7.8	Xe-133	5.245 d	7.3
Zr-97	16.9 hr	6.0	Te-132	3.258 d	6.9
Rh-105	1.473 d	5.4	Ce-143	1.375 d	5.9
Pd-109	13.46 hr	4.4	I-133	20.8 hr	5.9
Ce-134	1.375 d	4.3	Zr-97	16.9 hr	3.1
Mo-99	2.75 d	4.2	I-131	8.041 d	3.1
I-135	6.611 hr	4.2	Ba-140	12.79 d	2.9
Te-132	3.258 d	2.8	Xe-135	9.089 hr	1.9
Sr-91	9.5 hr	2.4	Pd-109	13.46 hr	1.4
Ru-105	4.439 hr	1.5	Ru-103	39.28 d	1.4
Xe-133	5.245 d	1.4	Ce-141	32.51 d	1.3
			Xe-133m	2.19 d	0.42
			Xe-131m	11.9 d	0.0058
10-DAY DECAY			**30-DAY DECAY**		
Radionuclide	**Half-Life**	**%**	**Radionuclide**	**Half-Life**	**%**
Xe-133	5.245 d	13.6	Ru-103	39.28 d	12.2
Ba-140	12.79 d	7.9	Ce-141	32.51 d	10.7
I-131	8.041 d	7.0	Ba-140	12.79 d	9.6
Mo-99	2.75 d	6.5	Zr-95	63.98 d	6.3
Te-132	3.258 d	6.2	I-131	8.041 d	4.5
Ru-103	39.28 d	4.8	Xe-133	5.245 d	3.5
Ce-141	31.51 d	4.6	Nd-147	11.06 d	3.4
Nd-147	11.06 d	3.3	Ce-144	284.3 d	1.4
Zr-95	63.98 d	2.2	Ru-106	1.008 yr	1.4
Rh-105	1.473 d	1.2	Te-129m	33.6 d	0.39
Sb-127	3.85 d	0.76	Te-132	3.258 d	0.32
Ce-143	1.375 d	0.69	Eu-156	15.19 d	0.29
Xe-133M	2.19 d	0.23	Xe-131m	11.9 d	0.13
Xe--131m	11.9d	0.05			

Table 2

EXAMPLES OF SOURCE TERMS
FROM 1 kT DETONATIONS
(One-day Post Detonation)

	Half-Life	BQ($x10^{15}$)	%Abundance
Mo-99	2.75 d	15.3	4.2
Ba-140	12.75 d	3.16	0.87
Xe-135	9.10	42.9	11.8
Xe-133	5.243 d	5.03	1.4

990

CONCENTRATION IF DILUTED BY EARTH'S ATMOSPHERE
COMPARED WITH DETECTION SENSITIVITIES

	Half-Life	μBq/m^3 on Dilution	Detection Sensitivity, μBq
Mo-99	2.75 d	3600	1
Ba-140	12.75 d	740	1
Xe-135	9.10 hr	10000	20
Xe-133	5.243 d	1200	20

Table 3

XENON-133 AND XENON-135 QUANTITIES AND RATIOS
VERSUS TIME AFTER A 1kT NUCLEAR DETONATION
10^{14}Bq

(Where XENON Gas Venting Occurs Within
5 Minutes After Detonation)

	Half-Life	5 Minutes	4 Hrs	12 Hrs	1 day	3 days	5 days	7 days
Xe-133	5.243 d	0.260	0.254	0.243	0.227	0.175	0.134	0.103
Xe-135	9.10 hr	114	84.6	46.0	18.4	0.477	0.0123	0.000319
Ratio	Xe-135/ Xe-133	440	333	189	81.2	2.72	0.092	0.0031

FIGURE 1

Particulate Radionuclide Analyzer

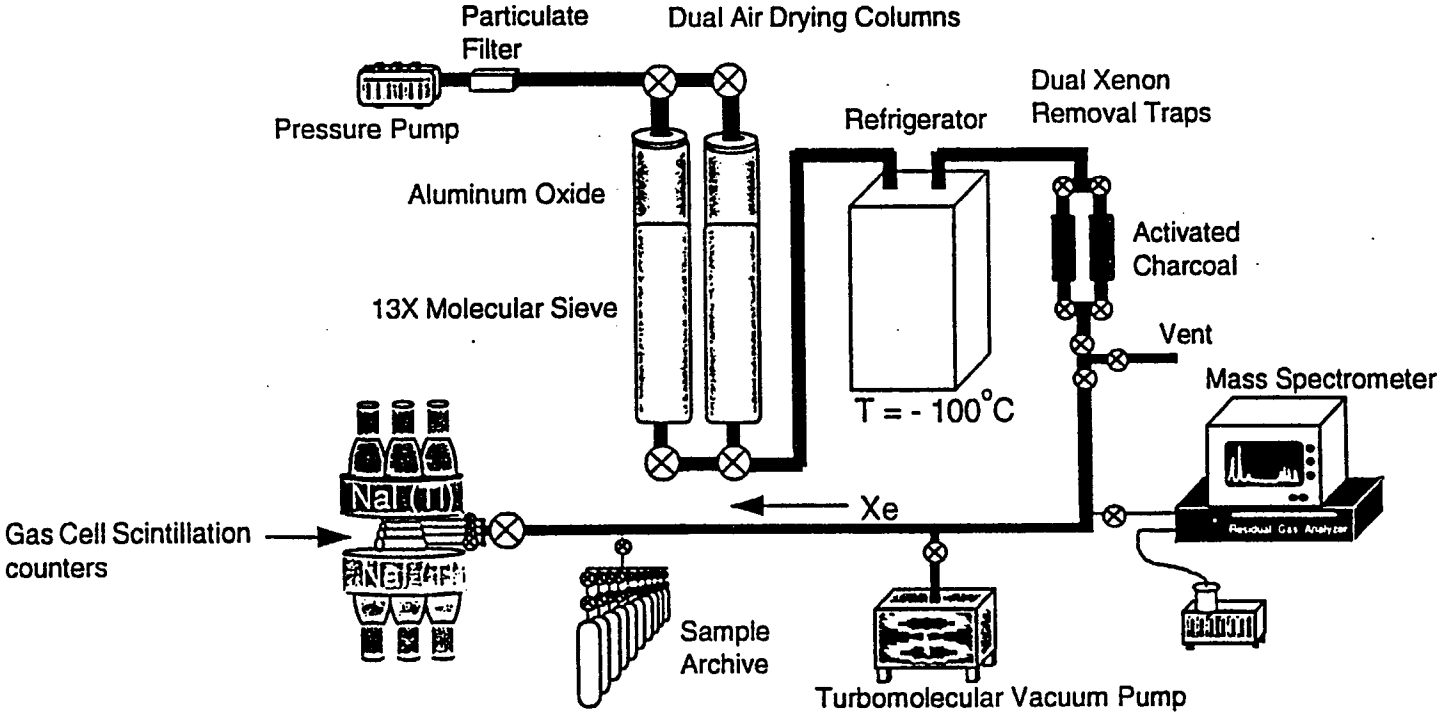
DOE/AFTAC Joint Field Test: McClellan AFB

- FULLY CONTAINED SYSTEM 3 x 3 x 7 FEET MOUNTED IN A COVERED TRAILER (7 x 12 x 7 FEET HIGH)
- BEGAN A 6 MONTH OPERATIONAL TEST IN JULY
- SAMPLES, ANALYZES AND TRANSMITS DATA AUTOMATICALLY
- OPERATIONAL CONTROLS, STATE-OF-HEALTH AND SOFTWARE UPGRADES AVAILABLE BY PHONE-IN

FIGURE 2

Xenon Radionuclide Analyzer

NEAR REAL-TIME ANALYSES OF THE XENON RADIONUCLIDES (ALL OPERATIONS AUTOMATIC)

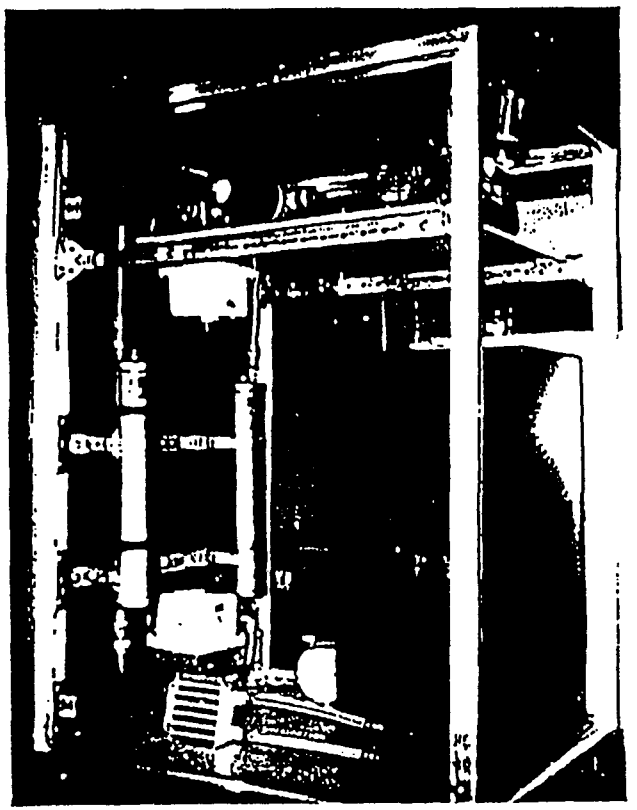


- **Designed to continuously separate xenon from the atmosphere at very high flow rates (40 m^3 of air per 8-hour collection period) which provides up to 50 fold greater sensitivity than other reported systems performance.**
- **Employs 8 hour collections versus one to two day collection periods which allow a 5 to 10 fold greater sensitivity for ^{135}Xe (9.1 hr).**
- **Employs highly efficient and low background beta coincidence gamma-ray spectrometry which provides a 10 fold greater sensitivity than available with other systems.**
- **Will Automatically transmit the data to NDC for forwarding to IDC.**

Figure 3

Xenon Radionuclide Analyzer

Fieldable Fully Automatic Prototype
Near Realtime Analyzer



Under Construction For Test In January 1996

- Fully automated sample acquisition, analysis and data transmission
- Portable - 3' x 3' x 7'
- Low power < 2.5 kW power consumption
- 8-hour cycle time